

Time-resolved collapse and revival of the Kondo state near a quantum phase transition

Christoph Wetli,¹ Johann Kroha,^{2,3} Kristin Kliemt,⁴ Cornelius Krellner,⁴

Oliver Stockert,⁵ Hilbert von Löhneysen,⁶ and Manfred Fiebig¹

¹Department of Materials, ETH Zurich, 8093 Zurich, Switzerland

²Physikalisches Institut and Bethe Center for Theoretical Physics,
Universität Bonn, Nussallee 12, D-53115 Bonn, Germany

³Center for Correlated Matter, Zhejiang University, Hangzhou, Zhejiang 310058, China

⁴Physikalisches Institut, Goethe-Universität Frankfurt,
Max-von-Laue-Str. 1, 60438 Frankfurt, Germany

⁵Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany

⁶Institut für Festkörperphysik and Physikalisches Institut,
Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany

One of the most successful paradigms of many-body physics is the concept of quasiparticles: excitations in strongly interacting matter behaving like weakly interacting particles in free space. Quasiparticles in metals are very robust objects. Yet, when a system's ground state undergoes a qualitative change at a quantum critical point (QCP) [1], these quasiparticles can disintegrate and give way to an exotic quantum-fluid state of matter where the very notion of particles comprising the system breaks down. The nature of this breakdown is intensely debated [2–5], because the emergent quantum fluid dominates the material properties up to high temperature and might even be related to the occurrence of superconductivity in some compounds [6]. Here we control the resurgence of heavy-fermion quasiparticles out of a photoexcited nonequilibrium state and monitor their dynamics towards the QCP in a time-resolved experiment, supported by many-body calculations. A terahertz pulse transforms heavy fermions in $\text{CeCu}_{5.9}\text{Au}_{0.1}$ into light electrons. Under emission of a delayed, phase-coherent terahertz reflex the heavy-fermion state recovers, with a memory time 100 times longer than the coherence time typically associated with metals [7, 8]. The quasiparticle weight collapses towards the QCP, yet its formation temperature remains almost constant. This suggests a revised view of quantum criticality in between disintegration and preservation of the quasiparticle picture.

At a QCP the critical temperature of a second-order phase transition approaches zero. There, critical fluctuations are generated by the energy allowed by Heisenberg's uncertainty principle rather than thermal excitations, and exotic properties such as non-Fermi-liquid behaviour, frustrated magnetism or unconventional superconductivity may emerge. In fact, matter as we conventionally know it may disintegrate in the vicinity of the QCP because fundamental concepts of condensed-matter physics lose validity.

This is exemplified by the notion of the quasiparti-

cle. All across condensed-matter physics, coherent excitations of many-body states can be described as particle-like objects. It allows the description of even very complex correlations in a simple picture of weakly interacting particles. In metals, Landau fermionic quasiparticles of remarkable stability are enforced by the Pauli principle. Heavy-fermions are particularly interesting in this respect, as they are amenable to disintegration near a quantum phase transition (QPT) due to their heavy effective mass and low characteristic energy. In heavy-fermion compounds, the $4f$ magnetic moments localized on rare-earth ions in the crystal lattice are exchange-coupled to the electron spins residing in the conduction band [1]. The conduction electrons form singlets with the $4f$ moments below the so-called Kondo lattice temperature T_K^* , typically about 10 K. Thus, instead of establish-

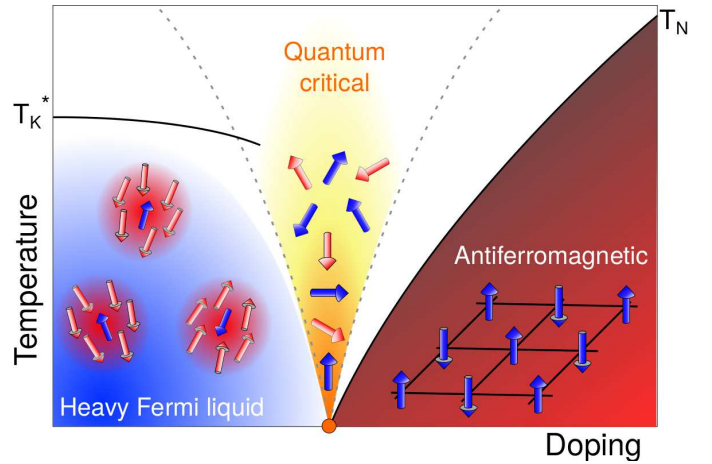


FIG. 1: **Heavy-fermion quasiparticles near a QCP.** We consider a general scenario, realized in $\text{CeCu}_{6-x}\text{Au}_x$, where the quasiparticles disintegrate near the QCP, giving way to an exotic state of matter, dominated by quantum fluctuations. In the quantum-critical region (yellow) the correlation length ξ of the quantum fluctuations is larger than the thermal length λ_{th} , that is, the distance a particle can travel quantum-coherently. Since $\lambda_{th} \propto 1/\sqrt{T}$ the system can counter-intuitively enter the quantum-critical region when the temperature is raised.

ing magnetic long-range order, a paramagnetic Fermi liquid phase is formed. This Kondo state [9, 10] is characterized by a sharp resonance in the $4f$ electron spectrum at the Fermi energy. Typically slightly below T_K^* , these resonances become lattice-coherent and form a narrow energy band of long-lived quasiparticles. Its flat dispersion indicates a $\sim 10^2$ times heavier effective mass than that of free electrons; hence the name “heavy fermion”. While in the traditional view of an antiferromagnetic QCP due to Hertz, Moriya and Millis [11–13] the heavy quasiparticles remain intact and support critical fluctuations of the magnetization, this picture is not valid in many materials [14].

$\text{CeCu}_{6-x}\text{Au}_x$ is one of the best-known heavy Fermi-liquid compounds of the latter type [1, 14–17]. Gold expands the CeCu_6 lattice. This leads to a relative increase of the RKKY interaction [18–20] between the $4f$ magnetic moments and induces a QPT to an incommensurate, antiferromagnetically ordered state at $x = 0.1$ [21, 22], see Fig. 1. Heavy fermions in $\text{CeCu}_{6-x}\text{Au}_x$ are thus an ideal system for studying the metamorphosis condensed matter may undergo near a QCP. A variety of experimental investigations by thermodynamic and transport measurements [22–24], neutron scattering [14, 15, 21, 25], and photoemission spectroscopy [8, 17] revealed valuable aspects of the phases and properties near a QCP. They did not access the coherent dynamics, however, so that a core issue of quantum matter research remains controversial: Does a fundamental concept of condensed-matter physics, such as the quasiparticle picture, remain valid near the QCP [11–13, 26], does it perish entirely [2, 3, 14], or can we extend the notion of Landau quasiparticles to a more general picture of critical quasiparticles [4, 5]?

In this work, we enter the realm of nonequilibrium dynamics to obtain direct information on the evolution of the quasiparticle picture towards the QCP in $\text{CeCu}_{6-x}\text{Au}_x$. We extinguish the Kondo state by irradiation with a THz pulse and monitor its time-resolved resurgence. The resulting THz echo pulse provides a direct measure for the spectral weight and for the Kondo temperature of the heavy-fermion state within a single experiment. Towards the QCP, the spectral weight collapses, reaching zero at 5 K. The Kondo energy scale, however, remains nearly constant at $T_K^* \simeq 7$ K. We thus observe a scenario of quasiparticle disintegration where the *time scale* on which quasiparticles form ($\sim 1/T_K^*$) remains finite, whereas the *probability* for quasiparticle formation (\sim spectral weight) as such collapses. Moreover, our experiment reveals a coherence time on the order of 10 picoseconds in metals where the coherence time is typically on the femtosecond timescale [7]. These results could change our established view of quantum criticality and allow first insights into its dynamical aspects.

We irradiate samples of $\text{CeCu}_{6-x}\text{Au}_x$ with the quantum-critical Au concentration $x = 0.1$ or with the antiferromagnetic concentration $x = 1$ with THz pulses in the frequency range of 0.3 – 3 THz. The pulses ex-

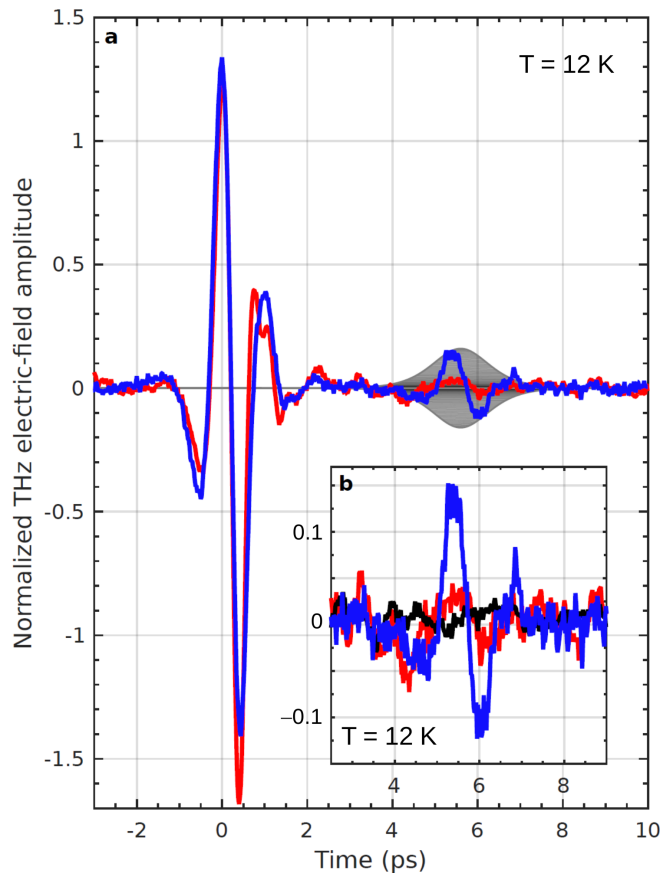


FIG. 2: **Time-resolved THz reflectivity of the heavy-fermion system $\text{CeCu}_{6-x}\text{Au}_x$.** Time dependence of the normalized THz electric-field amplitude for reflection from quantum-critical $\text{CeCu}_{5.9}\text{Au}_{0.1}$ (blue), from the antiferromagnetic compound CeCu_5Au (red) and from a platinum reference sample (black). Each time trace is normalized such that its entire integrated intensity is one (see Methods). The weight of the echo pulse in the $\text{CeCu}_{5.9}\text{Au}_{0.1}$ time traces exhibits a complex temperature dependence, while its delay time is essentially temperature independent, as analyzed in Fig. 3. The grey area shows the envelope of the time-delayed THz reflex of Eq. (2), fitted to the measured data.

cite electrons from the Kondo state to the conduction band but they do not ionize the system. We then record the reflected THz wave by electrooptical sampling (see Methods). The time traces in Fig. 2a exhibit an instantaneous reflex as response of the light conduction electrons. This is followed by a weaker “echo” pulse delayed by 5.8 ps. This echo is not to be confused with trivial reflex pulses from the THz generation crystal, cryostat windows etc. These artifacts were all identified at different delay times (see Methods and Supplement). Unlike these artifacts, the 5.8-ps echo does *not* appear on a Pt reference sample. It furthermore displays a complex temperature dependence, detailed in the following, that none of the trivial reflex pulses show.

Two features about this echo pulse are striking. First, its delay by 5.8 ps indicates quantum coherent dynamics

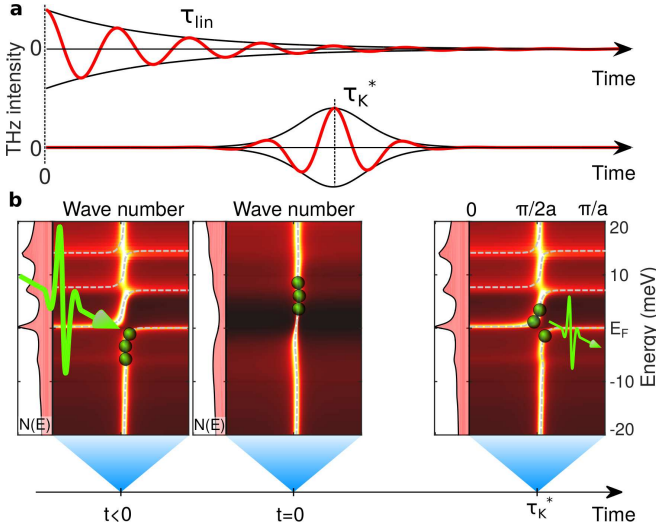


FIG. 3: Dynamics of heavy-fermion quasiparticle formation. **a** Sketch of the relaxation dynamics after photoexcitation of carriers into the conduction band. Top: immediate, exponential relaxation of the reemitted electric field resulting from a linear rate equation with constant relaxation rate $1/\tau_{lin}$, describing, e.g., exciton recombination in semiconductors. Bottom: solution (2) of the nonlinear rate equation (1). Here the heavy-fermion band is of many-body origin, i.e., its spectral weight and, hence, the relaxation rate are dynamically following the heavy-fermion band population. The pronounced echo pulse with delay time τ_K^* is an unambiguous signature of many-body Kondo physics. **b** Band structure of a multi-orbital Anderson-lattice model, calculated by nonequilibrium dynamical mean field theory (see Methods). Before the THz excitation ($t < 0$) the nearly flat heavy-fermion band near the Fermi energy $\varepsilon_F = 0$ accompanied by its crystal-field satellites at 8 and 13 meV, [15] are visible. Excitation with a THz pulse ($t = 0$) destroys the heavy-fermion state. The associated band collapses and all charge carriers become light electrons. After a time τ_K^* , resurgence of the heavy-fermion state occurs and the excess energy is released as time-delayed THz pulse.

of the THz-excited electrons on the order of 10 ps, about two orders of magnitude longer than the coherence time typically associated with photoexcited electrons in metals [7]. Second, the delayed response does not show instantaneous exponential decay, as one would expect from single-electron relaxation (Fig. 3a), but a compact pulse, well separated from the instantaneous reflex by a “dark time”.

The observed delay time $\tau_K^* \approx 5.8$ ps corresponds to a temperature $h/k_B\tau_K^* = 8.3$ K (with h and k_B Planck and Boltzmann constant, respectively), in good agreement with the Kondo temperature of $\text{CeCu}_{5.9}\text{Au}_{0.1}$ outside the quantum-critical region [15]. As seen in Figs. 2a and b, the echo is suppressed in the antiferromagnetic, non-heavy-fermion phase of CeCu_5Au [1, 21, 22], evidencing its Kondo-related origin: By the THz excitation, heavy electrons are excited instantaneously (within the THz pulse duration) from the heavy-fermion band

into the light part of the conduction band, as shown in Fig. 3b. This transition breaks the Kondo singlet and thus deletes the associated spectral weight of the heavy band. It then takes the coherence time τ_K^* to recreate this spectral weight, into which the excited electrons relax with emission of the initially absorbed energy as a delayed THz pulse. Because of energy-momentum conservation the THz excitation occurs only close to the avoided crossing of heavy and light conduction bands, split by the renormalized hybridization of order T_K^* , as depicted in Fig. 3b. The phase space for scattering of these THz-excited electrons is drastically reduced compared to optical excitation because of the low energy deposited by a THz photon. Hence the exceptionally long coherence time of ~ 10 ps is obtained in our experiment.

This THz dynamics is quantitatively explained by a simple rate-equation model. The electric-field amplitude $\bar{E}(t)$ of the emitted THz pulse is proportional to the temporal change of the density $\rho(t)$ of photoelectrons in the conduction band, or $\bar{E}(t) \sim d\rho(t)/dt = -\Gamma_{c \rightarrow hf} \rho(t)$, where $\Gamma_{c \rightarrow hf}$ is the transition rate of light conduction electrons to the heavy-fermion band. Via Fermi’s golden rule, $\Gamma_{c \rightarrow hf}$ is proportional to the spectral density of heavy-fermion states. The latter is proportional to the number of Kondo singlets present at time t , i.e., to the density of electrons residing in the heavy-fermion band, $\rho_0 - \rho(t)$. Since furthermore the dipole transition matrix element $d_{c \rightarrow hf}$ is related to the Kondo resonance width or the Kondo temperature via $k_B T_K^* \propto |d_{c \rightarrow hf}|^2$, one obtains an unusual *time-dependent* transition rate, namely $\Gamma_{c \rightarrow hf} = 2\pi(2k_B T_K^*/h)(\rho_0 - \rho(t))/\rho_0$. Thus, the rate equation for the normalized density of photoexcited electrons, $n(t) = \rho(t)/\rho_0$, reads

$$\frac{dn}{dt} = -\frac{4\pi}{\tau_K^*}(1-n)n. \quad (1)$$

Solving Eq. (1) yields the electric-field envelope of the THz reflex,

$$\bar{E}(t) = \frac{\bar{E}_0}{\cosh^2[2\pi(t/\tau_K^* - 1)]}, \quad (2)$$

where $\bar{E}_0 \propto \rho_0 |d_{c \rightarrow hf}|^2$. This envelope is shown in Fig. 2a and agrees well with our experimental observation. Note that both, the pulse delay and its temporal width, are controlled by the single parameter τ_K^* (Eq. (2)). The time-delayed revival of the signal is caused by the dynamical change of spectral weight in the final state of the relaxation process, expressed by the time-dependence of $\Gamma_{c \rightarrow hf}$. This is an unambiguous signature of correlated many-body dynamics. Noninteracting excitations like excitons would instead show instantaneous exponential relaxation at constant Γ (see Fig. 3a).

We now scrutinize the fate of the quasiparticles as the QCP is approached. Their spectral weight is obtained from the integrated, background-corrected intensity of the delayed THz reflex (see Methods). Even outside of the heavy Fermi-liquid phase, i.e., at high temperatures

or in the antiferromagnetic region, the system exhibits Kondo-like spin correlations which do not contribute to the coherent heavy-fermion quasiparticle weight but which do contain the Kondo scale $T_K^* = h/k_B\tau_K^*$, as reported earlier [17]. In our experiment these appear as time-delayed background features, faintly visible e.g. in the $x = 1$ sample in Fig. 2. They allow us to extract the delay time τ_K^* from the time traces even when the coherent heavy-fermion quasiparticle weight is vanishing.

Figure 4a shows that the heavy-fermion quasiparticle weight of the $\text{CeCu}_{5.9}\text{Au}_{0.1}$ sample rises below 150 K and down to about 25 K. As expected from the Kondo effect [10] and the thermally excited crystal-field states [15], this rise extends to remarkably high temperature and is logarithmic, see Fig. 4b. Below 25 K, however, the weight drops continuously, reaching zero at about 5 K (Fig. 4b). This constitutes the first direct, spectroscopic observation of quasiparticle disintegration near a QCP in any material. At the same time, the Kondo temperature, extracted from the pulse delay time and shown in Fig. 4c, drops only by about 5% and does not approach zero. Figure 4 thus reveals that while the quasiparticle spectral weight, i.e., the probability for the very existence of quasiparticles, collapses, there is, contrary to common belief, no change of the Kondo scale, i.e., the time it takes the quasiparticles to form ($\tau_K^* \propto 1/T_K^*$) remains nearly constant.

Our measurement is an unambiguous demonstration that these two aspects are not contradictory, as both the spectral weight and the pulse delay time are recorded here within one single THz reflectometry experiment. In fact, they are consistent with the observation of the so-called ω/T scaling [14] as an indicator of Kondo destruction. Our observations thus call for a revised view on heavy fermions near QCPs. In a wider perspective it also calls for a revised view of the concept of quasiparticles at quantum phase transitions in general.

Methods

Experiment. The $\text{CeCu}_{6-x}\text{Au}_x$ samples used in this study were cut from large, single crystals grown by the Czochralski technique. All samples were oriented by the x-ray Laue backscattering method, and faces perpendicular to the principal axes were polished using SiC. The specimens were mounted in a temperature-controlled Janis SVT-400 helium reservoir cryostat with Tsurupica windows. A Ti:Sapphire laser (800 nm, 130 fs, 1 kHz, 8 mJ/pulse) generated single-cycle THz pulses of a few nJ by optical rectification in a 0.5 mm ZnTe(110) single crystal. THz radiation with a spectral peak at 0.3–3 THz was incident onto the sample under 45° with the THz electric field parallel to its b axis. Because of the low THz pulse energy and the high THz reflectivity [23] of $\text{CeCu}_{6-x}\text{Au}_x$, heating by the THz pulses is negligible.

The electrooptical sampling was performed using the fundamental Ti:sapphire laser pulse as probe pulse that was time-delayed with respect to the reflected THz wave. The THz and probe beams were collinearly focused onto

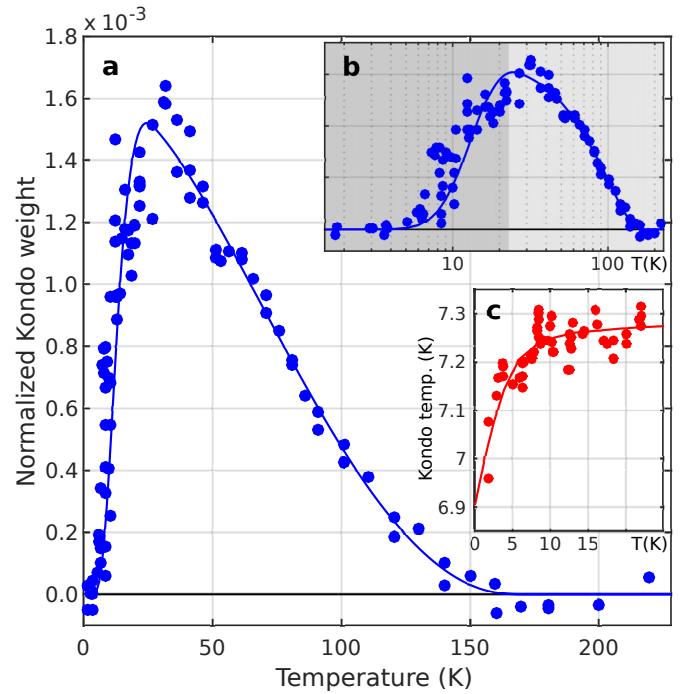


FIG. 4: **Evolution of the heavy-fermion quasiparticle in $\text{CeCu}_{5.9}\text{Au}_{0.1}$ towards the QCP.** **a**, Temperature dependence of the Kondo weight, extracted as described in Methods from the time-delayed THz “echo” pulse, seen e.g. in Fig. 2b. Disintegration of the quasiparticle weight below about 25 K is obvious. **b**, Data in (a) replotted for a logarithmic temperature scale. This reveals the logarithmic rise of the Kondo weight down to 25 K and its absence below 5 K. **c**, Kondo temperature extracted from the delay time of the THz “echo” pulse. With decreasing temperature, T_K^* exhibits a mere 5% drop. All lines are guides to the eye.

a ZnTe(110) crystal. The THz-induced ellipticity of the probe light is measured using a quarter-wave plate, a Wollaston polarizer and a balanced photodiode. Output from the latter was analyzed with a lock-in amplifier. In order to increase the accessible time delay between THz and probe pulses, Fabry-Perot resonances from the faces of the 0.5 mm short ZnTe(110) crystal were suppressed by extending the crystal with a 2.5 mm long THz-inactive optically bonded ZnTe(100) crystal.

Prior to the experiment we identified all the THz reflexes generated by the optical components in order to avoid confusion with the true THz signal generated in the $\text{CeCu}_{6-x}\text{Au}_x$. A measurement of all these THz artifacts is presented as a Supplement. The earliest of these artifacts appeared at a delay of 10 ps.

A set of data points from the electrooptical sampling represents the time trace of the THz field pulse between $t = -4$ ps and $t = +16$ ps. Each set was normalized by a factor such that its entire integrated intensity (the sum of the square of all the normalized data points in the set) equals one, i.e. all traces are scaled to the same over-all reflected power. Such normalized data are shown

in Fig. 2. They were used to derive the heavy-fermion quasiparticle weight as follows. The power of the echo pulse was calculated by integrating the squared electric field of the normalized time traces over the interval from 3 to 8 ps, where the echo pulse appears. These raw data exhibit an offset, caused by noise, the residual tail of the undelayed main pulse, and incoherent Kondo correlations [17], visible in our data as a temperature independent contribution above 150 K up to room temperature. Thus, this offset is not related to the Kondo quasiparticle weight. It was subtracted from the raw data to obtain the quasiparticle weight shown in Fig. 4a, b.

Theory. The dynamical band structure of the THz-excited heavy-fermion system, including the crystal-field satellite bands, was calculated using a nonequilibrium generalization of the dynamical mean-field the-

ory (DMFT) [27] for the multi-orbital Anderson lattice model with six local orbitals, grouped in three Kramers doublets. These orbitals represent the crystal-field-split $J = 5/2$ ground-state multiplet of the Ce 4f shell. An infinite onsite repulsion within the Ce 4f orbitals was assumed, enforcing the overall single-electron occupancy of the Ce 4f shell. As the impurity solver of the DMFT, an auxiliary-particle representation of the Ce 4f electron fields was employed within the Non-Crossing Approximation [16, 17, 28] (NCA), reaching down to base temperatures (before THz excitation) of $T \approx 0.1 T_K^*$. DMFT and NCA were generalized to the nonequilibrium case using the Keldysh technique [29]. The efficient implementation of the NCA algorithm of Ref. [28] was adapted for solving the DMFT with the multi-orbital NCA out of equilibrium.

-
- [1] v. Löhneysen, H., Rosch, A., Vojta, M. & Wölfle, P. Fermi-liquid instabilities at magnetic quantum phase transitions, *Rev. Mod. Phys.* **79**, 1015 (2007).
 - [2] Si, Q., Rabello, S., Ingersent, K. & Smith, J. L. Locally critical quantum phase transitions in strongly correlated metals, *Nature* **413**, 804 (2001).
 - [3] Coleman, P., Pépin, C., Si, Q. & Ramazashvili, R. How do Fermi liquids get heavy and die?, *J. Phys. Cond. Mat.* **13**, R723 (2001).
 - [4] Senthil, T., Vojta, M. & Sachdev, S. Weak magnetism and non-Fermi liquids near heavy-fermion critical points, *Phys. Rev. B* **69**, 035111 (2004).
 - [5] Wölfle, P. & Abrahams, E. Quasiparticles beyond the Fermi liquid and heavy fermion criticality, *Phys. Rev. B* **84**, 041101(R) (2011).
 - [6] Kenzelmann, M., Strässle, Th., Niedermayer, C., Sigrist, M., Padmanabhan, B., Zolliker, M., Bianchi, A. D., Movshovich, R., Bauer, E. D., Sarrao, J. L. & Thompson, J. D. Coupled superconducting and magnetic order in CeCoIn₅, *Science* **321**, 1652 (2008).
 - [7] Knoesel, E., Hotzel, A. & Wolf, M., Ultrafast dynamics of hot electrons and holes in copper: Excitation, energy relaxation, and transport effects, *Phys. Rev. Lett.* **57**, 12812 (1998).
 - [8] Kummer, K., Vyalikh, D. V., Rettig, L., Cortés, R., Kucherenko, Yu., Krellner, C., Geibel, C., Bovensiepen, U., Wolf, M., & Molodtsov, S. L., Ultrafast quasiparticle dynamics in the heavy-fermion compound YbRh₂Si₂, *Phys. Rev. B* **86**, 085139 (2012).
 - [9] Kondo, J. Resistance minimum in dilute magnetic alloys, *Prog. Theor. Phys.* **32**, 37 (1964).
 - [10] Hewson, A. C., *The Kondo Problem to Heavy Fermions*, Cambridge University Press, Cambridge (1993).
 - [11] Hertz, J. A. Quantum critical phenomena, *Phys. Rev. B* **14**, 1165 (1976).
 - [12] Moriya, T. *Spin fluctuations in itinerant electron magnetism* (Springer, Berlin, 1985).
 - [13] Millis, A. Effect of a nonzero temperature on quantum critical points in itinerant fermion systems, *Phys. Rev. B* **48**, 7183 (1993).
 - [14] Schröder, A., Aeppli, G., Coldea, R., Adams, M., Stockert O., v. Löhneysen, H., Bucher, E., Ramazashvili, R. & Coleman, P. Onset of antiferromagnetism in heavy-fermion metals, *Nature* **407**, 351 (2000).
 - [15] Stroka, B., Schröder, A., Trappmann, T., v. Löhneysen, H., Loewenhaupt, M. & Severing, A. Crystal-field excitations in the heavy-fermion alloys CeCu_{6-x}Au_x, *Z. Phys. B* **90**, 155 (1993).
 - [16] Ehm, D., Schmidt, S., Hüfner, S., Reinert, F., Kroha, J., Wölfle, P., Stockert, O., Geibel, C. & v. Löhneysen, H. High-resolution photoemission study on low- T_K Ce Systems: Kondo resonance, crystal field structures, and their temperature dependence *Phys. Rev. B* **76**, 045117 (2007).
 - [17] Klein, M., Nuber, A., Reinert, F., Kroha, J., Stockert, O. & v. Löhneysen, H. Signature of quantum criticality in photoemission spectroscopy at elevated temperature, *Phys. Rev. Lett.* **101**, 266404 (2008).
 - [18] Ruderman, M. A. & Kittel, C. Indirect exchange coupling of nuclear magnetic moments by conduction electrons, *Phys. Rev.* **96**, 99 (1954).
 - [19] Kasuya, T. A theory of metallic ferromagnetism and antiferromagnetism on Zeners model, *Prog. Theor. Phys.* **16**, 45 (1956).
 - [20] Yosida, K. Magnetic properties of Cu-Mn alloys, *Phys. Rev.* **106**, 893 (1957).
 - [21] Schröder, A., Lynn, J. W., Erwin, R. W., Loewenhaupt, M. & v. Löhneysen, H. Magnetic structure of the heavy fermion alloy CeCu_{5.5}Au_{0.5}, *Physica B* **199 & 200**, 47 (1994).
 - [22] v. Löhneysen, H., Sieck, M., Stockert, O. & Waffenschmidt, M. Investigation of non-Fermi-liquid behavior in CeCu_{6-x}Au_x *Physica B*, **223 & 224**, 471 (1996).
 - [23] Marabelli, F. & Wachter, P. Temperature dependence of the optical conductivity of the heavy-fermion system CeCu₆, *Phys. Rev. B* **42**, 3307 (1990).
 - [24] Pietrus T., Bogenberger, B., Mock, S., Sieck, M. & v. Löhneysen, H. Pressure dependence of the Néel temperature in antiferromagnetic CeCu_{6-x}Au_x for $0.3 \leq x \leq 1.3$, *Physica B* **206-207**, 317 (1995).
 - [25] Stockert, O., v. Löhneysen, H., Rosch, A., Pyka, N. & Loewenhaupt, M. Two-dimensional fluctuations at the quantum-critical point of CeCu_{6-x}Au_x, *Phys. Rev. Lett.* **80**, 5627 (1998).

- [26] Rosch, A., Schröder, A., Stockert, O. & v. Löhneysen, H. Mechanism for the non-Fermi-liquid behavior in $\text{CeCu}_{6-x}\text{Au}_x$, *Phys Rev. Lett.* **79**, 159 (1997).
- [27] Aoki, H., Tsuji, N., Eckstein, M., Kollar, M., Oka, T. & Werner, Ph. Nonequilibrium dynamical mean-field theory and its applications, *Rev. Mod. Phys.* **86**, 779 (2014).
- [28] Kroha, J. & Wölfle, P. Fermi and non-Fermi liquid behavior in quantum impurity systems: conserving slave boson theory, *Acta Phys. Pol. B* **29**, 3781 (1998).
- [29] Hettler, M. H., Kroha, J. & Hershfield, S. Nonequilibrium dynamics of the Anderson impurity model, *Phys. Rev. Lett.* **58**, 5649 (1998).

Acknowledgments. The authors are grateful for financial support by the SNSF via project No. 200021-14708 (M. F., C. W.) and by the DFG via SFB/TR 185 (J. K).

Author Contributions. All authors contributed to the discussion and interpretation of the experiment and to the completion of the manuscript. C. W. performed the experiment and the data analysis. O. S. and H. v. L. provided the $\text{CeCu}_{6-x}\text{Au}_x$ samples. K. K. and C. K. provided YbRh_2Si_2 samples for reference experiments. J. K. performed the theoretical analysis. J. K. and M. F. initiated the experiment and supervised the work. C. W., J. K. and M. F. drafted the manuscript.

Competing Interests. The authors declare that they have no competing financial interests.

Correspondence. Correspondence and requests for materials should be addressed to M. F. (email: manfred.fiebig@mat.ethz.ch) or J. K. (email: kroha@th.physik.uni-bonn.de).

Time-resolved collapse and revival of the Kondo state near a quantum phase transition

C. Wetli, J. Kroha, K. Kliemt, C. Krellner, O. Stockert, H. v. Löhneysen, M. Fiebig

Supplementary Information

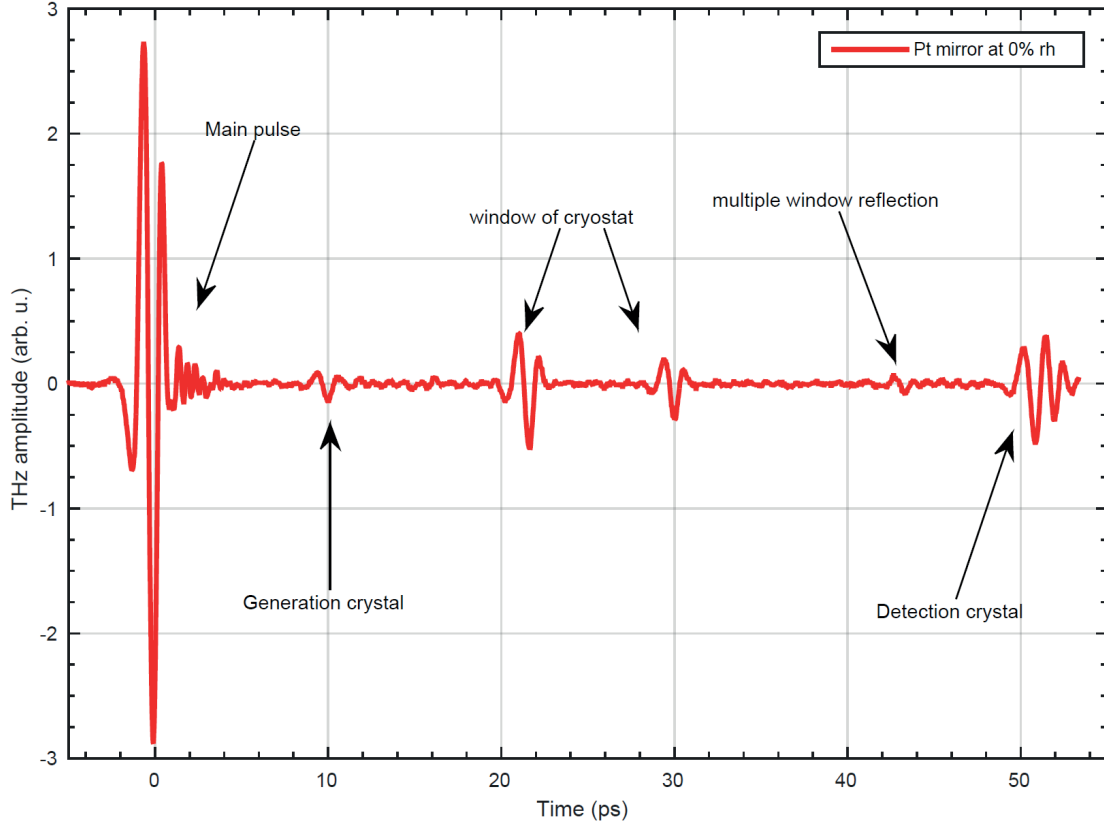


FIG. S1: THz response from a platinum mirror in the setup used for our experiments. Aside from the main pulse various reflex artifacts are observed whose origins are indicated in the plot. Note that no such artefact is present at a delay of 5.8 ps, the position of the Kondo echo pulse. Furthermore, the reflex artifacts in the plot exhibit none of the complex temperature dependence of the echo pulse depicted in Fig. 4 of the article.